

Observation of Spin Susceptibility Enhancement in the Possible FFLO State in CeCoIn₅

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(Dated: Version February 5, 2008)

We report ¹¹⁵In nuclear magnetic resonance (NMR) measurements of the heavy-fermion superconductor CeCoIn₅ in the vicinity of the superconducting critical field H_{c2} for a magnetic field applied perpendicular to the \hat{c} -axis. A possible inhomogeneous superconducting state, Fulde-Ferrel-Larkin-Ovchinnikov (FFLO), is stabilized in this part of the phase diagram. In 11 T applied magnetic field, we observe clear signatures of the two phase transitions: the higher temperature one to the homogeneous superconducting state and the lower temperature phase transition to a FFLO state. It is found that the spin susceptibility in the putative FFLO state is significantly enhanced as compared to the value in a homogeneous superconducting state. Implications of this finding for the nature of the low temperature phase are discussed.

PACS numbers: 74.70.Tx, 76.60.Cq, 74.25.Dw, 71.27.+a

The recently discovered heavy-fermion superconductor CeCoIn₅ with a quasi-2D electronic structure [1] offers a unique opportunity to investigate the interplay of unconventional superconductivity, magnetic fluctuations, quantum criticality, and non-Fermi liquid behavior. The experimental evidence indicates that CeCoIn₅ is an unconventional superconductor with, most likely, d -wave gap symmetry [2, 3, 4, 5] below $T_c = 2.3$ K. The H_{c2} transition in this compound becomes first-order below $\simeq 0.7$ K, for magnetic field (H) applied both parallel and perpendicular to the $(\hat{a}\hat{b})$ planes [2, 6, 7], signaling that this compound is a superconductor subject to the Pauli limit [8]. Thus, it offers for the first time the possibility to investigate the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) [9] state in an unconventional inorganic superconductor. This effect is related to the fact that in a spin singlet superconductor the destruction of superconductivity by a magnetic field can be accomplished in two ways. Cooper pairs may break up either because of the coupling of the spin degrees of freedom to the field (Pauli paramagnetism) or because of the effect of the field on the orbital degree of freedom (vortices). A novel FFLO phase is predicted to occur when Pauli pair-breaking dominates over the orbital effects [9]. In this new state the superconducting order parameter varies periodically in space and Cooper pairs acquire finite momentum, $|\vec{q}| \sim 2\mu_B H / \hbar v_F$. This concept, developed in the context of solid state physics, is now highly topical for ultra-cold atomic Fermi gases [10]. In addition, the study of the FFLO is of significant interest in elementary-particle physics [11].

Single crystal samples of CeCoIn₅ appear to be ideal material for a microscopic investigation of the FFLO state owing to their high purity, layered structure, and large spin susceptibility. Many experiments [12, 13, 14, 15] have identified a possible phase transition to a FFLO state. Even though these measurements identified the

phase transition within the Abrikosov superconducting (SC) state (henceforth referred to as uniform SC state) in the vicinity of the H_{c2} , the nature of this fundamentally new SC state remains to be elucidated.

Here we report on a study of ¹¹⁵In magnetic shift in CeCoIn₅ using nuclear magnetic resonance (NMR) in a magnetic field applied parallel to $(\hat{a}\hat{b})$ plane. The high field low temperature (T) part of the phase diagram was explored. The measurements probe the local spin susceptibility. At 11 T, they clearly reveal two phase transitions, at T_c and T^* as indicated in the inset to Fig. 1. We show that the phase transition at T_c is associated with the transition to the uniform SC state. We find that below T^* the spin susceptibility is enhanced compared to the value in a uniform SC state, but reduced compared to the normal state. Within the phase we did not observe any normal state regions, interpreted as evidence for the emergence of a spatially inhomogeneous SC state [15]. Thus, we argue that the state below T^* may be either a FFLO state that cannot simply be viewed as a stack of SC and normal regions, or a state with a different spin fluctuation character, and consequently with possibly different order parameter compared to the uniform SC state.

Single crystals of CeCoIn₅ were grown by a flux method as described in Ref. [1]. The crystal structure of CeCoIn₅ is tetragonal and consists of alternating layers of CeIn₃ and CeIn₂. This leads to two inequivalent ¹¹⁵In sites per unit cell [4, 16]. Our sample is believed to be of a good quality since all observed In ($I = 9/2$) and Co ($I = 7/2$) lines are narrow (in the normal state FWHM for Co line is $\simeq 12$ kHz and $\simeq 20$ kHz for In). Furthermore, the quality of the sample is confirmed by the observed position of the nine satellite lines for both In sites, which are in agreement with the previously reported values [4]. From the position of the satellites we infer that no

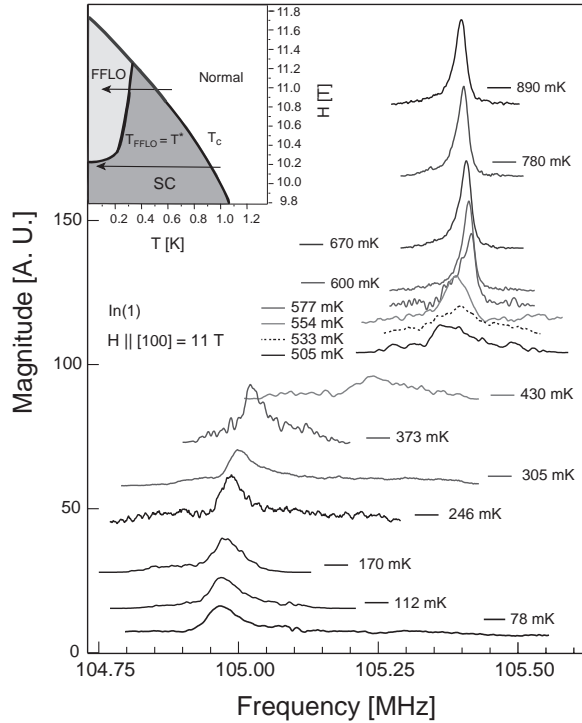


FIG. 1: NMR spectra of $^{115}\text{In}(1)$ (central transition) as a function of temperature at 11 T magnetic field applied in the [100] direction. The spectra are normalized to their corresponding areas and offset for clarity. The offset is proportional to the corresponding T . Inset: Sketch of the $H - T$ phase diagram of CeCoIn_5 for $H \perp \hat{c}$ [12]. Arrows indicate the fields in which spectra discussed in the paper were taken.

significant structural changes take place down to 70 mK and that H was aligned to better than 2° and 4° with respect to the sample's $(\hat{a}\hat{b})$ plane and \hat{a} axis, respectively.

The shift measurements of the axially symmetric $^{115}\text{In}(1)$ site reported here were made on a large single crystal of CeCoIn_5 . The NMR spectra were recorded using a custom built NMR spectrometer and obtained, at each given value of H , from the sum of spin-echo Fourier transforms recorded at each 10 (or 20) KHz interval. The shift was determined by the diagonalization of the nuclear spin Hamiltonian including the quadrupolar effects. Temperature independent orbital contribution ($\simeq 0.13\%$) to the shift was subtracted. Below T_c the demagnetization field lowers the observed local field at the nucleus site. Therefore, an accurate determination of the spin shift may be precluded by this shielding field and the values of the extracted shift could be underestimated. The shift data presented in this paper correspond to a lower bound of the intrinsic spin susceptibility below T_c . However, for H parallel to $(\hat{a}\hat{b})$ planes the demagnetization factor is relatively small.

The low T environment was provided by a $^3\text{He}/^4\text{He}$ dilution refrigerator. The RF coil was mounted into the mixing chamber of the refrigerator, ensuring good thermal contact. To be able to span over a wide frequency range, we used the 'top-tuning' scheme in which

the variable tuning delay line and matching capacitor were mounted outside the NMR probe. For H values in the vicinity of 11 T the central transitions of the two In sites are separated by only $\simeq 400 \text{ KHz}$, which is of the order of the shift of the $\text{In}(1)$ line at T^* . Therefore, it was necessary to check the satellite frequencies to assure that the correct site is followed as a function of T . The sample was field-cooled. In order to avoid heating of the sample by RF pulses we used an RF excitation power much weaker than usual and repetition times of the order of a tenth of a second to several seconds, depending on T .

In Fig. 1 spectra of $^{115}\text{In}(1)$ as a function of T at $H = 11 \text{ T}$ are displayed. Two phase transitions are evident. The first transition is from the normal state to the uniform SC state at $T_c \simeq 550 \text{ mK}$ and the second is from the uniform SC state to the supposed FFLO state between $370 < T < 430 \text{ mK}$. As T_c is crossed a severe loss of the signal intensity is observed. The intensity in the SC state at $T \simeq 500 \text{ mK}$ is reduced by an order of magnitude with respect to the normal state intensity. The loss of signal intensity is due to RF shielding by the superconducting currents. This observation confirms that below $\simeq 550 \text{ mK}$ the sample is indeed in a SC state.

We will discuss the possible microscopic nature of the low T phase. The lineshapes in the low T phase are clearly distinct from the ones at higher temperatures. Nonetheless, these lineshapes are not consistent with a traditional vortex lattice field distribution [17]. That is, the spectra are broaden well beyond the expectations for the vortex lattice lineshapes. The progressive loss of the signal intensity (corrected for the Boltzmann factor), due to the extra shielding of RF, ceases on lowering the T below 370 mK. However, the signal intensity still remains significantly lower compared to the normal state intensity. This would imply that below $T \simeq 370 \text{ mK}$ the SC order parameter, whatever its nature might be, is fully developed.

For $370 < T < 470 \text{ mK}$ the signal is extremely weak, almost indiscernible from the noise, as evident in Fig. 1. We point out that the weakness of the signal is not only due to RF penetration problems. The main cause of the loss of signal intensity is an enhancement of the spin-spin decoherence rate, T_2^{-1} . A fast decoherence rate (shorter than the dead time for the signal detection, $\simeq 6 \mu\text{s}$) implies strong enhancement of a magnetic fluctuation component parallel to H on approaching T^* . It is well known that enhanced fluctuations are precursory to magnetic transitions or transitions associated with vortex dynamics. However, the transition at T^* cannot be associated with the changes in vortex dynamics, since vortices must be in a solid state, *i.e.* static on the NMR time scale, at all temperatures below T_c , otherwise the problem of RF penetration would not be encountered throughout this T range. Any static rearrangement of vortices at a fixed value of the applied field cannot account for the observed shift variation [17]. There is evidence of abundant antiferromagnetic spin fluctuations in CeCoIn_5 and of

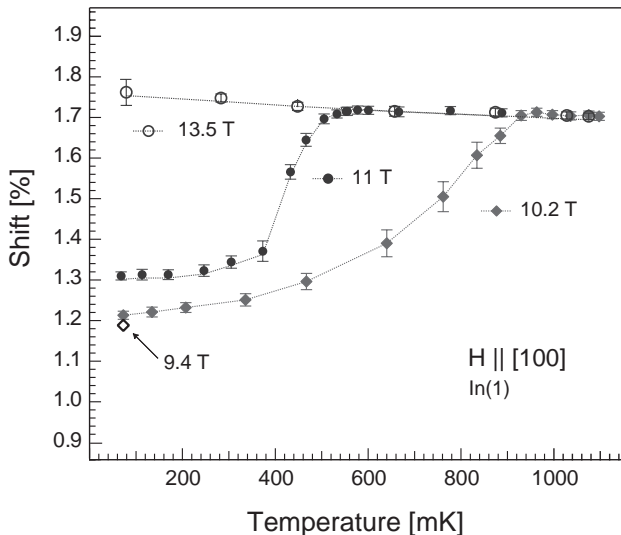


FIG. 2: The magnetic shift of $^{115}\text{In}(1)$ as a function of T at 10.2, 11, and 13.5 T. The dotted lines are guide to the eye. The open diamond is the shift at 9.4 T.

their coexistence with uniform superconductivity [18, 19]. Therefore, it is conceivable to think that magnetic fluctuations change from antiferromagnetic to ferromagnetic-like or that the nature of antiferromagnetic fluctuations changes [20] at T^* . In the former scenario, this in turn could cause the changes of the SC order parameter.

To gain further insight into the nature of the FFLO state, we proceed to the analysis of the spin shift. In Fig. 2 the T dependence of the shift at 10.2, 11, and 13.5 T is shown. In the high field normal state there is no evidence of any phase transition. While the 11 T data exhibits two phase transitions, our 10.2 T data reveals that the sample undergoes one phase transition to the uniform SC state. In the uniform SC state at both 10.2 and 11 T the decreasing shift with decreasing T reveals a well known suppression of the spin susceptibility consistent with spin-singlet pairing [3, 4]. Below T^* at 11 T, the T dependence of the shift is strongly reduced. Overall the spin shift seems to decrease faster on approaching T^* as compared to the its T dependence in the uniform SC state. However, this difference is not very significant on the reduced T scale, as illustrated in the following paragraph.

In Fig. 3, the temperature dependence of the normalized shift (spin susceptibility) in the SC states is plotted. In the uniform SC state the temperature dependence of the local magnetization, which is proportional to the shift, is expected to be quadratic, when $\mu_B B > k_B T$, with the residual $T = 0$ value due to the field induced shift in the spin-split density of states [21, 22]. The solid line in Fig. 3 indicates such a T dependence. The data in the uniform SC state at both fields is in agreement with the quadratic T dependence. However, below T^* there is an obvious discrepancy, well outside the error bars, between data at 11 T and the expected T dependence in the uniform SC state. The discrepancy is particularly

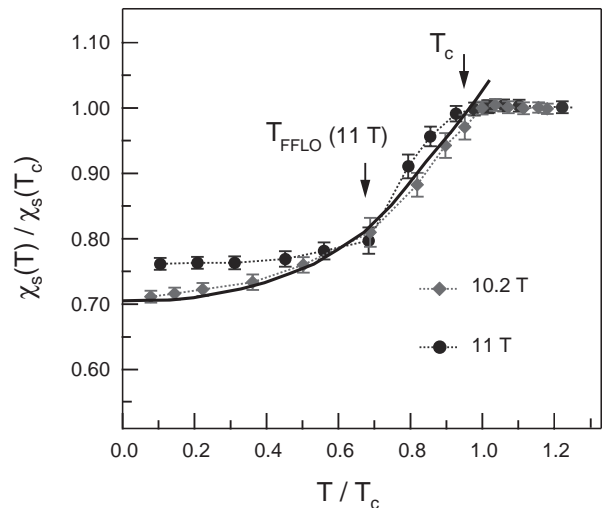


FIG. 3: Normalized $^{115}\text{In}(1)$ shift in the SC states as a function of T at 10.2 and 11 T. Solid line denotes the quadratic T dependence. Transition temperatures (T_c and $T_{\text{FFLO}} \equiv T^*$) are adopted from Ref. [12] for comparison.

pronounced at very low temperatures, *i.e.* $T \rightarrow 0$ limit.

Therefore, we conclude that in the state below T^* the spin shift is enhanced compared to the value in a uniform SC state, but reduced compared to the normal state. The enhancement is not a trivial effect due to increase of the applied field by 0.8 T on χ , but it is a consequence of intrinsically different nature of the FFLO phase as compared to the uniform SC phase. This is because in a uniform SC state at 70 mK decreasing H from 10.2 T by the same amount of 0.8 T changes the shift by only $\simeq 0.024\%$. This is 4 times less than the observed enhancement between 10.2 T and 11 T, as shown in Fig. 2.

Below T^* , we do not observe any signal at frequencies corresponding to the normal state signal. Therefore, a low T state in our sample cannot simply be viewed as a stack of spatially well separated SC and normal regions, static on the NMR time scale. However, this does not rule out the FFLO nature of the low T phase. The spin shift can be enhanced by a more complex periodic superposition of SC and “normal” regions, defined by a spectrum of spin-polarized quasiparticles [23]. Indeed, calculations indicate that a finite quasiparticle density of states (DOS) is induced at the Fermi level in the FFLO state [23, 24]. This finite DOS enhances the spin susceptibility, since $\chi \propto \text{DOS}(E_F)$. Furthermore, the χ is found to be T independent in the FFLO phase for T below $\sim \frac{1}{2}T^*$ [21, 24], in agreement with our data. As a matter of fact, our measurements are consistent with two possible pictures of the FFLO state: static and dynamic. In the former, the FFLO state can be viewed as a static periodic modulation of the SC and “normal” regions, in which case the data imposes an important constraint on the local magnetization of the “normal” quasiparticle regions. Mainly, it must be *significantly lower than the magnetization of the normal state*, since no signal is observed at frequencies corresponding to the normal state

signal. Alternatively in a dynamic scenario, the modulation of the amplitude of the order parameter in the FFLO state can fluctuate in space over a distance of the order of $1/q$. Thus, a measured shift would represent a volume weighted average of the shifts in the SC and “normal” regions. In such a state, the NMR shift is higher than in the uniform SC state and no distinct signal from the normal regions should be observed, consistent with our measurements.

The observed enhancement of the spin shift could be induced by the appearance of a small static component (ferromagnetic or canted antiferromagnetic) in the spin degrees of freedom. However, there is no evidence from our high field data (at 13.5 T in Fig. 2) that a magnetic phase transition connected with an extension of the FFLO phase takes place in the normal state. Therefore, the existence of the magnetic moment requires pair coherence. In a more exotic scenario, the low T phase could be a mixture of singlet and triplet pairs. It is possible that in high magnetic field it becomes energetically favorable to form triplets through the ferromagnetic coupling channel [25], even though the states of mixed parity are not *a priori* allowed in a material with inversion center. Finally, χ may also be enhanced by the uncondensed electrons that coexist with the SC ones on a microscopic lengthscale, so that no separate signal is observed at the frequencies corresponding to the normal state signal. The existence of uncondensed electrons could be explained within the context of multiband SC [26].

There are evident discrepancies between our data and the results reported in Ref. [15]. Besides the lack of the signal at frequencies corresponding to the normal state signal, the relative shift changes are inconsistent. The normal state $^{115}\text{In}(1)$ shift [27] presented here is comparable to the shifts reported in Ref. [4, 15]. However, the relative shift changes between the low temperature

SC and normal state, evident in Fig. 2, are $\simeq 10$ times larger than those reported in Ref. [15]. Furthermore, the transition temperatures found in this work are in agreement with specific heat measurements of Bianchi *et al.* [12] and different from those observed in Ref. [15]. It is unlikely that the sample quality is the origin of these discrepancies, since the spectral lineshapes and their corresponding widths in high T normal state and at the lowest T are comparable. It is possible that differences stem from the exact sample alignment with respect to H [13].

In conclusion, our NMR study of $\text{In}(1)$ magnetic shift in CeCoIn_5 in a magnetic field applied parallel to SC planes has confirmed the existence of two phase transitions at low T in the vicinity of H_{c2} . At 11 T, we show that the phase transition at T_c clearly corresponds to a transition to the uniform spin-singlet SC state. We find that the main feature of the low T state below T^* is the enhancement of the spin susceptibility as compared to the value in a uniform SC state. In the FFLO scenario for the low T state, our data supports two possible pictures. First, the static FFLO state, in which the local magnetization of the “normal” regions must be significantly lower than the magnetization of the normal state. Second, the dynamic FFLO state in which the modulation of the amplitude of the order parameter fluctuates in space. Alternatively, it is possible that the low T phase arises from an intricate interplay of magnetism and superconductivity.

We are very grateful to V. Mineev, Č. Petrović, A. Benlagra, A. B. Vorontsov, M. J. Graf, J. P. Brison, S. Krämer, and A. J. Sauls for helpful discussions. Special thanks to W. P. Halperin for the magnet time and useful discussions. This work was supported by the funds from Brown University, and the Grenoble High Magnetic Field Laboratory, under European Community contract RITA-CT-2003-505474.

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